Research on the method of measuring the activity of new α -particle targeted therapy radiopharmaceutical Ac-225 based on CdZnTe detector

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Abstract: Ac-225 is a radionuclide with significant potential for clinical application in α-particle targeted tumour therapy. Precise quantification of its activity is essential for therapeutic application. This research used a CdZnTe detector to measure the γ energy spectrum of a Th-229 solution, intending to investigate the factors affecting measurement outcomes and the efficiency calibration method for activity measurement techniques applicable to Ac-225 clinical treatment.The results indicate that only the Full-energy peak generated by the 440.46 keV γ ray emitted by Bi-213 is suitable for analyzing the activity of Ac-225. The emissivity of γ-rays emitted by Ac-225 and its daughter nuclide varies significantly across different databases, with a maximum discrepancy of 36.8%; thus, it is essential to adjust the emissivity while computing the findings. The calibration of CdZnTe detection efficiency using the Eu-152 analogue source method and the analogue calculation method yields deviations of 2.0% and -4.6% from the reference values, respectively, indicating that both calibration methods provide reliable measurement results. The impact of the cascade conformity effect of γ-ray on CdZnTe measurement findings is negligible. However, its effect on HPGe measurement results can be as high as 32.4%. The research findings serve as a reference for developing a technical method to measure

the activity of the radiopharmaceutical Ac-225 applying CdZnTe.

Key words: Ac-225, CdZnTe, Efficiency calibration, Full-energy peak, Emissivity, Cascade conformity effect.

1. Introduction

The novel α -particle-targeted therapy can damage tumour cells when the radiopharmaceutical Ac-225 decays, and it can be used for the treatment of leukaemia, non-Hodgkin's lymphoma, malignant melanoma, bladder cancer, glioma, neuroendocrine tumours, and prostate cancer. [1-4] At present, North America and Europe have conducted a significant amount of clinical research. [5-8]. Ac-225 can be extracted from Th-229 by radiochemical method or produced by accelerator. [9-10]. Radioactivity is often used in nuclear medicine to measure the dose of radiopharmaceuticals. Usually, the activity of radiopharmaceuticals is measured by an activity meter, and a Standard Source needs to establish the linear relationship between activity and current. Ac-225 is a newly developed radiopharmaceutical, and the activity cannot be measured by the current commonly used activity meter. According to the decay properties of Ac-225, the radioactivity of Ac-225 can be measured by the α energy spectrum method, the β counting method, and the γ energy spectrum method. [11-15]. When using α-energy spectrometry and β-counting, sample pre-treatment is more complicated, which is not favourable for clinical use. γ -energy spectrometry is simple to take samples and can record the energy spectrum of γ-rays emitted by radionuclides to obtain more information about the samples to be tested and to identify the radioactive impurities in the drug. Activity meters measure the activity of radiopharmaceuticals by measuring the average cumulative current, which cannot differentiate between nuclides, and the accuracy of the measurement results for low-activity samples is poor. However, gamma spectrometry can improve the accuracy of the measurement results for low-activity samples by increasing the measurement time. Therefore, using γ-energy spectrometry to measure the activity of radiopharmaceuticals can improve the quality of their clinical applications.

The HPGe γ energy spectrometer has good energy resolution, but the detector needs to be at liquid nitrogen temperature to work correctly. The equipment is

challenging to maintain, expensive, and bulky, so it is difficult to use widely in the clinic. Sodium iodide, lanthanum bromide, and other scintillation γ spectrometers have relatively poor energy resolution. cdZnTe detectors have good energy resolution of γ-rays, the equipment is lightweight. It can be operated at room temperature, the crystal volume is small, and the detection efficiency of γ-rays is low, which is suitable for the measurement of the activities of high-activity radiopharmaceuticals and has a good advantage in the field of the activity measurement technology of radiopharmaceuticals. The efficiency calibration of the γ energy spectrum detection system is a key step in the measurement of radionuclide activity in the sample using the γ energy spectrum method, which has a direct impact on the measurement results, and the methods of efficiency calibration include the Standard Source method, the monoenergetic nuclide efficiency curve method, the simulation source method, the simulation calculation method, etc. The efficiency calibration method can measure the radionuclide activity in the sample using the γ energy spectrum method, simulated source method, simulation calculation method, etc. [16] The Ac-225 Standard Source and the monoenergetic γ nuclide Standard Source, which can be used for the CdZnTe γ energy spectrometer efficiency calibration, require high activity, which is difficult to obtain, and can only be used with the analogue source method and analogue computation method to scale the efficiency. When analyzing the γ energy spectrum, in addition to the detection efficiency, factors such as the choice of γ-ray Full-energy peak, the γ-ray emissivity, and the cascade conformity effect impact the analysis results.

Several investigations [11-15] documented the measurement of Th-229 or Ac-225 utilising HPGe, nevertheless they did not examine the many factors influencing the outcomes. Additionally, there are limited reports on the measurement of Th-229 or Ac-225 employing CdZnTe. This study aimed to quantify a Th-229 solution in radioactive equilibrium utilising a CdZnTe γ-energy spectroscopy detector. We investigated the impact of various factors, including efficiency calibration methods, selection of full-energy peaks, γ-ray emissivity, and cascade conformity effects, on the analytical results for Ac-225. The objective was to establish a methodological

foundation for measuring the activity of the radiopharmaceutical Ac-225 with a CdZnTe detector.

2. Principle and Methods

The simulated Standard Source emited γ-rays with energy comparable to that of the γ-rays released by the nuclide under examination, allowing the efficiency value of the γ-rays from the simulated source to directly substitute the efficiency of the γ-rays from the nuclide being evaluated. Despite the similarity in energy between the nuclide under examination and the γ-rays emitted by the simulated Standard Source, the emission processes may differ. If the γ-rays from the simulated Standard Source exhibit a more pronounced conformal effect, the efficiency value of the scale will experience considerable deviation, which can be reduced or potentially eliminate by increasing the distance (SD) from the sample to the detector surface[17-18].

The detector's efficiency for γ-ray detection is calculated applying the following equation:

$$\varepsilon(E) = \frac{N}{TA\eta K} \qquad (1)$$

Where: ε(E): the Full-energy peak efficiency for γ-rays with energy E;

N: net area of γ-ray Full-energy peak with energy E.

T: Measurement of the live time.

A: activity of Standard Source used by efficiency calibration.

η: emissivity of γ-rays of energy E emitted by Standard Source.

K: activity decay correction factor for Standard Source.

In the absence of Standard Source, detection efficiencies can also be obtained using Monte Carlo methods to accurately simulate the sample and measurement system.

3.Equipment and materials

3.1 Gamma energy spectrum detector

The CdZnTe γ spectrometer used in the experiments is the DT-01C11005 10× 10 × 5mm manufactured by Dietech3Quasi-hemispherical CdZnTe detector, for

Cs-137 emitted energy of 662 keV γ-rays with energy resolution better than 1.5%, custom energy spectrum analysis software.

The HPGe γ energy spectrometer is a BE5030 planar HPGe detector from Canberra, with a crystal size of Φ81 × 31 mm, a nominal relative efficiency of 50%, and an energy resolution of 1.87 keV (601332 keV for Co), and the γ energy spectrum analysis software is Genie2000.

3.2 Efficiency calculation software

The detection efficiency of the CdZnTe γ-detection system was simulated using MCNP5, and the detection efficiency of the HPGe γ-detection system was simulated and calculated using Canberra's LABSOCS software.

3.3 Standard Source and Samples

China Tongfu Co., Ltd. conducted the experiments applied in the [152]Eu simulation standard, with the uncertainty of nuclide-specific activity not exceeding 2.5% (k=2). The British National Physics Laboratory produces the Th-229 Standard Source. The nominal activity is 1062 Bq, and a 10 mL solution is contained in ampoules with a diameter of 15 mm.

4. Experiments and results

4.1 Radioactive balance

To evaluate the γ energy spectrum of Ac-225, the Th-229 standard solution produced in 2013 was used, and the decay diagram of Th-229 is illustrated in Figure 1.[19]
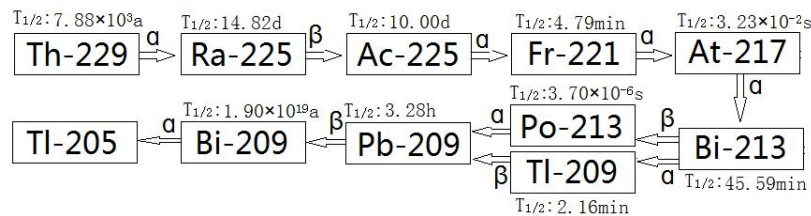

Fig. 1 decay diagram of th-229

According to the decay diagram of Th-229, except Bi-209, the longest half-life of the decaying daughter nuclide is 14.82 days. The Th-229 solution used in the experiment was produced 10 years ago, so the Th-229 and daughter nuclide reached a radioactive balance, and Ac-225 decayed according to the half-life of the mother

nucleus so that it could be used for a long time. According to the following research results, because the gamma rays emitted by Ac-225 are not available, we can only use the gamma rays emitted by daughter nuclide to analyze its activity. When measuring Ac-225 by gamma-ray spectrometry, only the nuclides before Pb-209 need to reach radioactive balance, and the half-life of Bi-213 is the largest, which is 45.59min, so it takes 5 hours to measure Ac-225.[20]The activity of Ac-225 can only be analyzed by gamma rays emitted by the daughter nuclide after the nuclides before Pb-209 reaches radioactive balance.

4.2 Gamma ray Full-energy peak

Table 1 lists the gamma rays emitted by Th-229 and daughter nuclide with energy greater than 40 keV and emissivity greater than 1%.[21] Fig. 2 shows the gamma spectrum obtained by measuring the Th-229 solution with a CdZnTe detector, and Fig. 3 shows the gamma spectrum obtained by measuring the Th-229 solution with an HPGe detector.

Table 1 Main γ -rays emitted by TH-229 and daughter nuclide

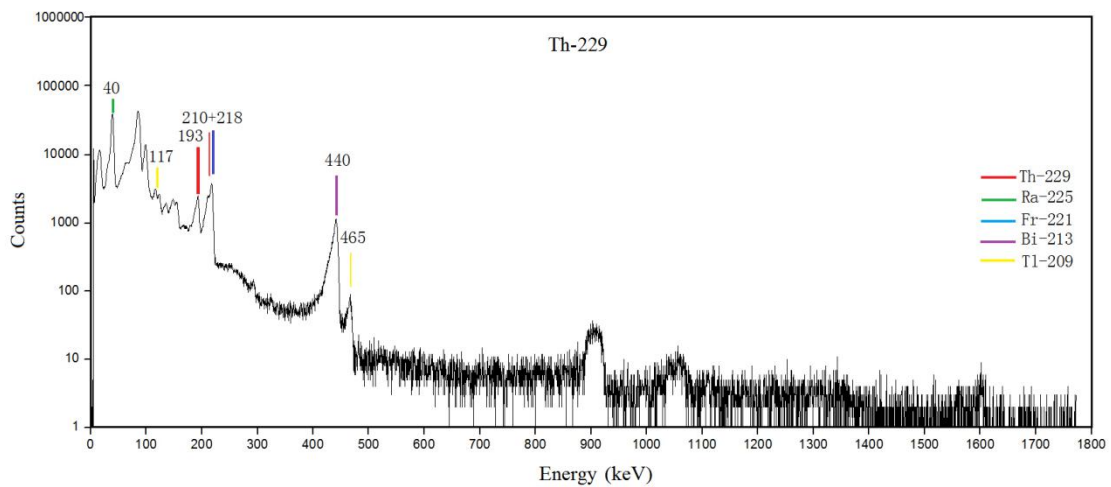| Nuclide	Gamma ray energy (keV)	Emissivity (%)	Nuclide	Gamma ray energy (keV)	Emissivity (%)
Th-229	86.25	1.33	Ac-225	99.91	1.01
	86.40	2.57	Fr-221	218.19	11.60
	136.99	1.18	Bi-213	440.46	26.10
	156.41	1.19	Tl-209	117.21	84.30
	193.51	4.41		465.13	96.90
	210.85	2.80		1567.09	99.80
Ra-225	40.00	30.00	--	--	--

Fig. 2 γ energy spectrum of Th-229 solution measured by CdZnTe detector
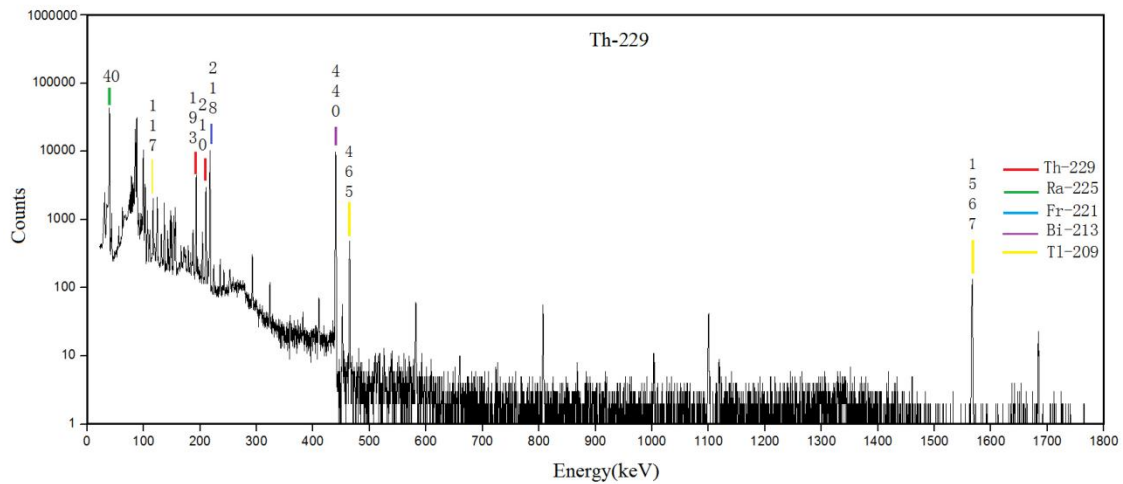
Fig. 3 γ energy spectrum of Th-229 solution measured by HPGe detector

When Th-229 and its daughters decay, they produce a lot of X-rays and gamma rays. There are 103 rays with an emission rate greater than 0.1%[22] and 13 gamma rays with an emission rate greater than 1%. The measured γ-energy spectra show that the γ-energy spectra below 300 keV are very complex, with many Full-energy peaks superimposed on each other and interfering with each other, resulting in a significant bias in the calculation of the Full-energy peak counts and emissivities.

When analyzing the γ-energy spectrum, choose γ-rays with large emissivities, no overlapping Full-energy peaks, and little interference around them. By analyzing the γ energy spectra measured by HPGe, the eight γ rays in Fig. 3 can be used as the activity calculations of Th-229 and its daughter nuclide.

Due to the relatively large FWHM (half height width) of the CdZnTe measured γ-rays Full-energy peak with low-energy trailing, it is more difficult to separate the γ-rays with similar energies Full-energy peak in the spectrum. The process of calculating the peak area is relatively complicated. The peak area of the CdZnTe measured γ-energy spectrum in this work is obtained by numerical fitting, which will be presented in another paper. According to the measured γ-energy spectrum of Th-229 solution with CdZnTe, although the γ-rays at 218.19 keV have a higher emissivity, the Full-energy peak overlaps and interferes with the Full-energy peak of 210.85 keV, 193.51 keV, and other γ-rays in this energy interval, which results in the distortion of the peak shape and a more significant error of the peak area calculated by

the spectral analysis software. The peak area error may be even more significant for for the lower energy γ-rays, superimposed on the interference of X-rays. The detection efficiency of CdZnTe for high-energy γ-rays is extremely low, so the Full-energy peak of 1567.09 keV cannot be seen in the γ-energy spectrum, and the count rate of the Full-energy peak of 465.13 keV is also very low. Therefore, when using CdZnTe, only the 440.46 keV γ-ray emitted by Bi-213 can be used to analyze the activity of Th-229 or Ac-225.

4.3 Emissivity

Th-229 and its daughter nuclide emit γ-rays with more energy during decay. Table 2 lists the primary γ-ray emissivity data in the nuclear databases of CENDL, ENSDF, CEA, and JENDL, as well as the data in the nuclear databases that come with the γ-energy spectrometry analysis software Genie2000, and there is no Th-229 and its daughter nuclide related emissivity data in commercial γ-energy spectrometry analyzers such as Gamma Vision. 229 and its daughter nuclide-related nuclear data need to be edited by the user. Table 3 lists the branching ratio data of Bi-213 decays from different nuclear databases.

Table 2 γ-ray emissivity of Th-229 and its daughter nuclide from different nuclear databases

Nuclide	Energy (keV)	Emission rate (%)				
		CENDL [21]	ENSDF [23]	CEA [19]	JENDL#[24]	Genie2000
Th-229	193.51	4.41±0.06	4.3±0.3	--*	4.41	4.41
	210.85	2.80±0.40	2.7±0.3	--	2.77	2.8
Ra-225	40.00	30.00±0.70	30.00±0.70	30.00±0.70	30.00	--
Fr-221	218.19	11.60±0.30	12.57±0.30	11.42±0.15	11.4	10.9
Bi-213	440.46	26.10±0.30	25.8±0.3	26.10±0.30	25.8	16.5
Tl-209	117.21	84.30±0.20	77.62±0.95	77.22±0.27	75.7	--
	465.13	96.90±0.20	96.19±0.97	96.62±0.05	95.3	--
	1567.09	99.80±0.20	99.53±0.57	99.71±0.05	--	--

*: missing data; #: uncertainty of data not given.

Table 3 Branching ratios of Bi-213 decays for different nuclear databases

Parent nuclide	Daughter nuclide	Branching ratio (%)			
		CENDL[#21]	ENSDF [23]	CEA[#19]	JENDL [24]
Bi-213	Po-213	97.91	97.86±0.10	97.91	97.86±0.1
	Tl-209	2.09	2.14±0.11	2.09	2.14±0.1

\#: Uncertainties of the data are not given.

From the data in Tables 2 and 3, most of the γ-ray emissivity and branching ratios of Bi-213 decays in the four databases are similar. However, the emissivity data of the two larger emissivity, 218.19 keV and 440.46 keV γ-rays, vary greatly. The emissivity data of 218.19 keV γ-rays in Genie2000 has a deviation of 13.3%, the emissive data of 440.46 keV γ-rays in Genie2000 has a deviation of 36.8% concerning the data in CENDL, and the emissive data of 117.21 keV γ-rays in JENDL has a deviation of 10.2% concerning the data in CENDL. Therefore, corrections to the emissivity are required when analyzing Th-229 or Ac-225 activity using the γ energy spectrum analysis software. See 4.4 for recommended nuclear data.

4.4 Detection efficiency
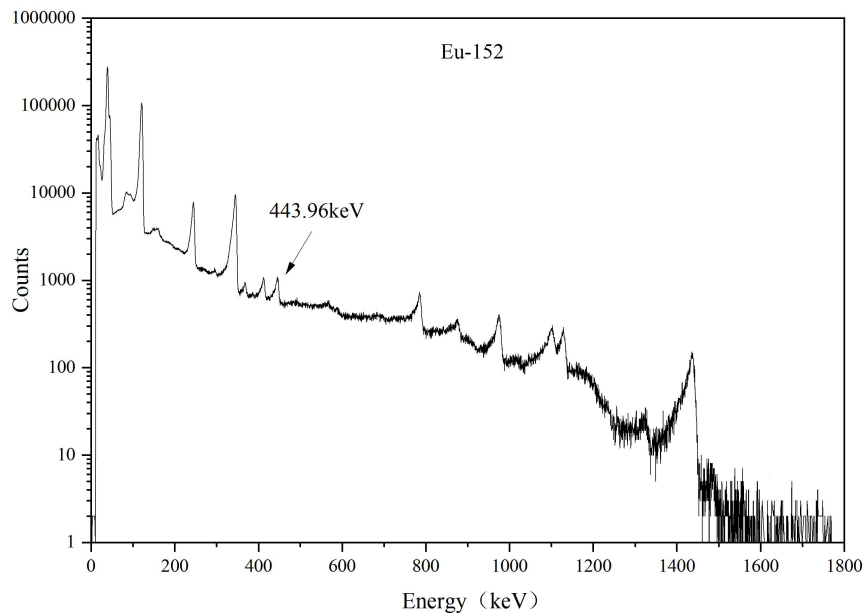### 4.4.1 Analogue source method

Fig. 4 Eu-152 γ energy spectrum measured by CdZnTe

CdZnTe relative to HPGe detection efficiency energy resolution is poor, the

detection efficiency is low, and the commonly used HPGe efficiency calibration Standard Source is unsuitable for CdZnTe. Eu-152 emits a γ-ray with an energy of 443.96 keV (emissivity of 3.1%), no interference peaks in the range of ± 30 keV in the Full-energy peak position, and the maximum emissivity of Ac-225 daughter nuclide is 440.46 keV γ-rays. 440.46 keV γ-ray energy can be used as an analogue source, and Figure 4 shows the Eu-152 γ energy spectrum measured using CdZnTe. In the laboratory, an Eu-152 analogue source was made by filling 10 mL of Eu-152 Standard Source (activity of 4365 Bq) in the same ampoule as Th-229 Standard Source. Table 4 shows the Ac-225 activities obtained using the CdZnTe, HPGe detector and Eu-152 analogue source efficiency calibration method, and the CdZnTe and HPGe detector γ-ray Full-energy peak efficiencies for 443.96 keV γ are also labelled in Figure 5.

Table 4 Ac-225 activities obtained by the efficiency calibration method using analog sources

Detectors	Measured parameters	Full-energy peak efficiency	Ac-225 activity (Bq)	Activity uncertainty	Relative Deviation from Nominal activity
HPGe	SD=2mm	0.01985	1278	3.0%	20.3%
HPGe	SD=102mm	0.00293	1026	3.0%	-3.4%
CdZnTe	Same calculation model	0.00146	1083	3.5%	2.0%

From the measurement results in Table 4, the deviation between HPGe and Standard Source is 25.3% when SD=2mm and only -3.4% when SD=102mm. The main reason is that the gamma rays of 443.96 kev emitted by Eu-152 can cascade with gamma rays of 964.08 keV and 1085.87 keV, which reduces the Full-energy peak count. When SD=2, the probability of cascade coincidence is high, and when SD reaches 102mm, the probability of cascade coincidence is minimal, and the influence on the results can be ignored. Therefore, when Eu-152 is used to simulate the calibration efficiency of the source method, the distance between the sample and

HPGe should be more than 102 mm.

The detection efficiency of CdZnTe for high-energy gamma rays is very low. When measuring the gamma spectrum of Eu-152, the probability of 443.96 kev gamma rays cascading with 964.08keV, 1085.87keV, and other gamma rays is extremely low, and the deviation between the results obtained by using Eu-152 as a simulation Standard Source is 2.0%. Therefore, Eu-152 is reliable as the simulation Standard Source for measuring activity by Ac-225 γ -ray spectrometry.

4.4.2 Simulation calculation method

The simulation calculation method is convenient for obtaining the detection efficiency curve of γ -the ray spectrum measurement system. In this work, the Monte Carlo particle transport platform MCNP5 is used to simulate and calculate 10× 10× 5 mm.3Detection efficiency of quasi-hemispherical CdZnTe detector for 10mL solution in φ Φ15mm ampoule. In the calculation, the ampoule with Standard Source is placed vertically on the horizontal table, and the CdZnTe is placed horizontally on the vertical line of the ampoule. The distance between the detector's and ampoule surfaces is 2mm (SD=2mm). The efficiency diagram calculated using the ideal model of characterized CdZnTe [25-27] is shown in Figure 5.

LabSOCS, a passive efficiency calibration software developed by Canberra Company, and the characterization file of the detector, the detection efficiency of HPGe for 10mL solution in φ Φ15mm ampoules was simulated and calculated. During calculation, the ampoule bottle is vertically placed in the center of the HPGe detector, and there is a protective cover with a thickness of 2mm between the ampoule bottle and the detector. In order to study the possible cascade conformity effect of γ -rays emitted by Ac-225 and daughter nuclide, the efficiency curve when SD is 102mm is also calculated, and the calculation results are shown in Figure 5.
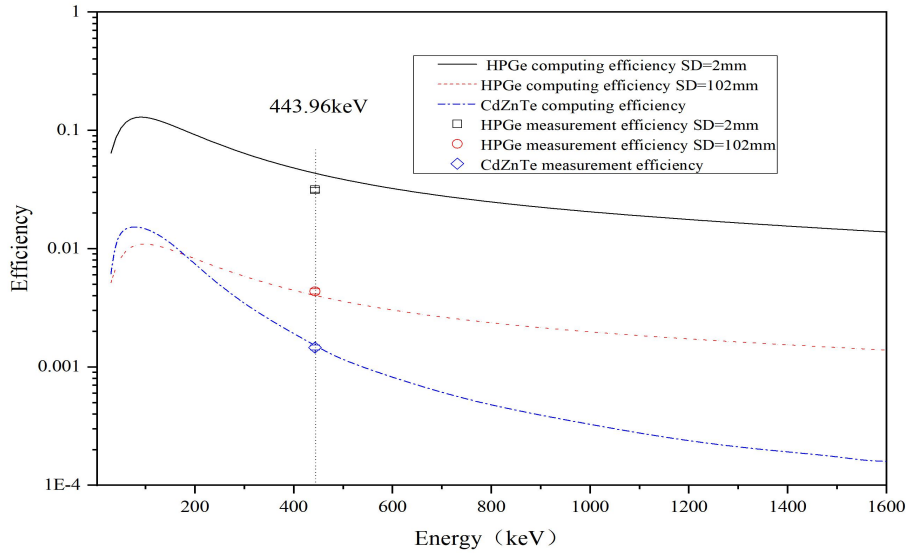
Fig. 5 Simulation calculation and measurement results of detection efficiency

The calculation results in Fig. 5 show that the detection efficiency of CdZnTe and HPGe for low-energy γ-rays is high. The detection efficiency increases gradually with the increase of energy before about 100 keV, and the detection efficiency will be smaller with the increase of energy after the energy of the γ-rays is more than 100 keV.The change in the detection efficiency of CdZnTe is swift with the change of energy, and the change in the detection efficiency of HPGe is much smoother than that of CdZnTe. The detection efficiency of HPGe for 59.5 keV γ-rays is 4.0 and 5.7 times higher than that of 662 keV and 1000 keV γ-rays, respectively, while that of CdZnTe for 59.5 keV γ-rays is 23.3 and 47.8 times higher than that of 662 keV and 1000 keV γ-rays, respectively, and even higher than that of 1500 keV γ-rays. 1500 keV γ-ray detection efficiency is nearly two orders of magnitude larger.

The activity of Th-229 and its daughter nuclide inside the ampoule was analyzed using the calculated HPGe efficiency curves. Table 5 shows the analysis results under SD=2mm and SD=102mm conditions. For the calculation of the results, the emissivity of each γ-ray was calculated using data from the JENDL database, except for the γ-ray of 1567.09 keV. The uncertainties in the activity were considered only for the uncertainty in the calculated value of the efficiency and the uncertainty in the Full-energy peak counts, and the uncertainty in the emissivity was not calculated.

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Table 5 Results of Th-229 and its daughter nuclide activity measured by HPGe under different SD conditions

Nuclide	Energy (keV)	SD=2mm			SD=102mm		
		Activity (Bq)	Activity uncertainty	Relative Deviation from Nominal activity	Activity (Bq)	Activity Uncertainty	Relative deviation from Nominal activity
Th-229	193.51	1023	1.2% -3.7	-3.7%	1061	1.2% -0.1%	-0.1 percent
	210.85	1095	3.1%	12.5%	1201	3.1%	13.1%
Fr-221	218.19	1052	2.0% - 1.0%	-1.0%	1084	2.0% 2.0% 2.0% 2.0% 2.0% 2.0% 2.0	2.1%
Bi-213	440.46	1020	1.2% 2.1% Bi-213 440.46 1020	-3.9%	1035	1.2%	-2.5%
Tl-209	117.21	769	4.8%	-27.6%	1034	4.8%	-2.6%
	465.13	724	3.8%	-31.8%	996	3.8%	-6.2%
	1567.09	718	3.6%	-32.4%	1009	3.6%	-5.1%

The analytical results in Table 5 indicate that when the standard deviation is 102mm, the calculated results of 117.21keV, 117.21keV and 117.21keV γ-rays emitted by Tl-209 have a significant deviation, reaching 32.4% at the maximum, which is due to the cascade coincidence of the three γ-rays emitted by Tl-209. The closer the Standard Source is to the detector, the greater the influence of the coincidence effect on the results, and vice versa. However, when the SD is 102mm, there is still a maximum deviation of 6.2%, which may have some influence. If the activity of Th-229 or Ac-225 is to be analyzed by gamma rays with these three energies, it must be measured under more extensive SD conditions. However, due to the limited height of the lead chamber of HPGe in the laboratory, this work has not been measured under more significant SD conditions.

In addition to the above three gamma rays, the calculation results of 40.00 kev gamma rays emitted by Ra-225 are also quite different, mainly caused by the slight deviation between the geometric model and the actual situation in the simulation

calculation. This slight deviation has little influence on higher-energy gamma rays but greatly influences low-energy gamma rays. When the simulated efficiency curve is used to analyze the activity of Th-229 or Ac-225, 40.00keV γ -rays can be omitted.

The maximum deviation of the calculation results of 210.85keV γ -ray emitted by Th-229 is 13.1%, which the deviation of emissivity may cause. The emissivity of 210.85keV γ -ray in four nuclear databases is in the range of 2.7-2.8, but the value in reference [28] is 3.25, and the deviation between the results obtained from this emissive data and Nominal activity is 3.6%, so the emissive of 210.85keV γ -ray is 3.25, which may be more reliable.

The results obtained applying γ-rays with energies of 193.51 keV, 218.19 keV, and 440.46 keV all agree with Nominal activity, proving that the simulated calculated efficiencies in the range of these energy regions are reliable.

MCNP5 simulation was used to calculate the CdZnTe detection efficiency, and the radioactivity of Th-229 and its daughter in the ampoule was also analyzed. The results are shown in Table 6.

Table 6 Results of Th-229 and its daughter nuclide obtained by CdZnTe simulation calculation efficiency

Energy (keV)	Full-energy peak net count rate (n/s)	Full-energy peak efficiency	Activity (Bq)	Activity uncertainty	Relative Deviation from Nominal activity
440.46	0.4026	0.00154	1013	3.5%	-4.6%

From the results in Table 7, the deviation of the measured results from Nominal activity obtained using the simulated efficiency curves is -4.6% and the method is reliable.

5.Conclusion

When applying commercial software to analyse the gamma radiation spectrum of the Ac-225 solution, it is essential to verify the emissivity data in the database and amend it if required. Both the simulated source method and the simulation calculation method yield dependable detection efficiency; however, when employing HPGe to

measure Ac-225, it is essential to elevate the Eu-152 simulation standard source to mitigate the impact of the cascade conformity effect on the measurement results. Conversely, the influence of the cascade conformity effect can be disregarded when utilising CdZnTe. CdZnTe exhibits inferior energy resolution compared to HPGe, and a low-energy tail characterises the full-energy peak. CdZnTe exhibits inferior energy resolution compared to HPGe, and the full-energy peak displays a low-energy tail. The calculation of the Full-energy peak area is intricate, necessitating the selection of a Full-energy peak with minimal or no interference to provide precise findings. In analysing the γ-energy spectrum of CdZnTe, it may be pertinent to assess the activity of Ac-225 solely through the 440.46 keV γ-ray full-energy peak.

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